

Landau level evolution driven by band hybridization in mirror symmetry broken ABA-stacked trilayer graphene

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Layer stacking and crystal lattice symmetry play important roles in the band structure and the Landau levels of multilayer graphene. ABA-stacked trilayer graphene possesses mirror-symmetry-protected monolayer-like and bilayer-like band structures. Broken mirror symmetry by a perpendicular electric field therefore induces hybridization between these bands and various quantum Hall phases emerge. We experimentally explore the evolution of Landau levels in ABA-stacked trilayer graphene under electric field. We observe a variety of valley and orbital dependent Landau level evolutions. These evolutions are qualitatively well explained by considering the hybridization between multiple Landau levels possessing close Landau level indices and the hybridization between every third Landau level orbitals due to the trigonal warping effect. These observations are consistent with numerical calculations. The combination of experimental and numerical analysis thus reveals the entire picture of Landau level evolutions decomposed into the monolayer- and bilayer-like band contributions in ABA-stacked trilayer graphene.

Recent progress on the study of quantum Hall effect (QHE) in graphene revealed novel physics such as half-integer QHE in monolayer graphene [1, 2], spin-valley quantum Hall ferromagnetism [3], spin phase transition and quantum spin Hall state at charge neutrality point [4, 5], and Hofstadter's butterfly in graphene/h-BN superlattice [6–8].

Graphene has a unique band structure and related properties drastically depending on layer number, stacking, and symmetry. Monolayer graphene (MLG), AB-stacked bilayer graphene (BLG) and ABC-stacked trilayer graphene (TLG) possess linear, parabolic, and cubic band dispersions, respectively. These systems have inversion symmetry and breaking inversion symmetry results in band gap formation. For the case of multilayer graphene, the inversion symmetry of the system can be controlled with perpendicular electric field [9–13]. In these multilayer systems, the integer and fractional quantum Hall phase transitions are also tunable with a perpendicular electric field [5, 14–21].

Trilayer graphene can have a different kind of stacking, called ABA-stacking (Fig. 1(a) left). ABA-stacked TLG has mirror symmetry. Reflecting this mirror symmetry, ABA-stacked TLG possesses a combined MLG-like and BLG-like band structure. The MLG-like band corresponds to mirror-symmetric wave function, whereas the BLG-like band corresponds to mirror-antisymmetric wave function. These two bands can hybridize when mirror symmetry is broken. In previous works, the mirror symmetry of the system is controlled by a perpendicular electric field [25–28]. Therefore, ABA-stacked TLG is a

novel platform to investigate the effect of band hybridization on the QHE. Under a perpendicular electric field, ABA-stacked TLG is expected to show complex but rich Landau level (LL) evolution, such as LL crossings and valley splitting due to the LL index dependent hybridization processes [29–31]. These features, however, have not yet been well revealed. In gapped bilayer graphene, it has been shown that the trigonal warping effect results in three fold degenerate LLs at low magnetic field and small carrier density regime [32]. In mirror symmetry broken ABA-stacked trilayer graphene, it is also expected that the trigonal warping affects LL orbital hybridization to induce anti-crossings between specific LLs [31]. At such anti-crossing points, the LL orbital states are tunable by an electric field; this may allow for further study on many-body phenomena such as quantum Hall ferromagnetism and fractional quantum Hall effect. Previous studies on the QHE in ABA-stacked TLG have revealed LL crossings due to the existence of independent MLG-like and BLG-like bands and unconventional sequence of the quantum Hall plateaus; which is explained by taking into account the all hopping parameters γ_0 to γ_5 (Fig. 1(a) left) and the onsite energy difference δ between A1, B2, A3 and B1, A2, B3 [23, 24, 26, 28, 33]. On the other hand, the perpendicular electric field dependence of the QHE in ABA-stacked TLG remained experimentally unclear, though the emergence or stabilization of a few novel quantum Hall plateaus have been observed [26, 28].

In this study, we reveal detailed picture of the evolution of the LL structure in ABA-stacked TLG under a perpendicular electric field. Importantly, all tight-binding

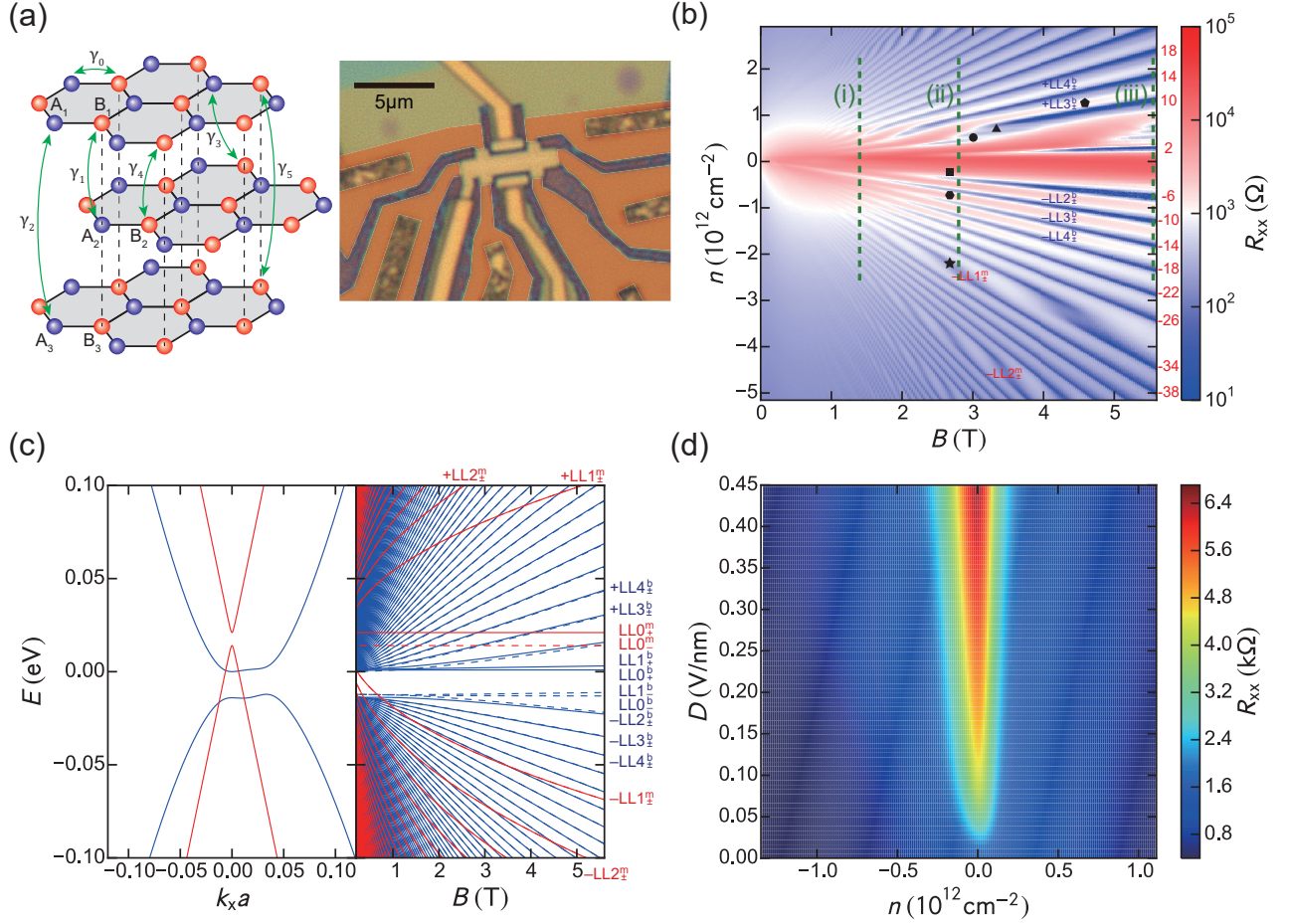


FIG. 1 (a) Left : Schematic image of the ABA-stacked trilayer graphene with tight-binding parameters [22]. Right : Optical microscope image of the TLG device encapsulated between h-BN layers. The device was shaped by reactive ion etching. The areas highlighted in red are unetched and used as contact leads. (b) Carrier density n and magnetic field B dependence of longitudinal resistance R_{xx} measured at $D = 0$ V/nm. The numbers in red on the right side of the figure show the filling factor at each local minimum of R_{xx} . (c) Numerical calculation of a typical band structure of ABA-stacked TLG around K point (left figure) and B dependence of the Landau levels at $D = 0$ V/nm (right figure). This calculation is performed using the tight-binding Hamiltonian with all tight-binding parameters [22–24]. The band structure is plotted as a function of $k_x a$ for $k_y a = 0$ where (k_x, k_y) is the wave number vector measured from K point and $a = 0.246$ nm is the lattice constant. The red (blue) colored lines show the MLG-like (BLG-like) bands at $B = 0$ T and their evolutions into the Landau levels. The solid (broken) lines in the right figure indicate the valley index K (K'). (d) n and D dependence of R_{xx} measured at 10 K. Gate capacitances are estimated so that the R_{xx} peak lies along $n = 0$.

parameters, γ_0 to γ_5 and δ , which describe the nature of the bulk graphite, play important roles in the LL structures of the ABA-stacked TLG as well as the onsite energy differences induced by an electric field. By applying perpendicular electric field, we find series of LL crossings and anti-crossings which originate from the hybridized MLG-like and BLG-like bands by breaking the mirror symmetry. We also observe anti-crossing structures mediated by the trigonal warping effect, which hybridizes LLs with the LL index difference of integer times 3, for both electron and hole carriers. These behaviors are well

reproduced by numerical calculations based on the tight-binding model with the MLG-like and BLG-like LL orbital basis, involving all tight-binding parameters γ_0 to γ_5 , δ and average onsite energy differences Δ_1 between the layers induced by the electric field.

TLG is encapsulated between h-BN layers in order to achieve a high carrier mobility and to apply a higher electric field than that for suspended devices (Fig. 1(a) right). Both graphene and h-BN were obtained via mechanical exfoliation. The number of graphene layers was confirmed optically via a contrast measurement. As de-

scribed below, the stacking of the TLG was confirmed as ABA from the measurement of the Landau fan diagram. We used pickup technique to fabricate the BN/TLG/BN stack and made the Ohmic contacts to graphene from the edge [34]. To control the carrier density (n) and the perpendicular displacement electric field (D) independently, we adopt a dual-gate structure [9]. All measurements were performed at 1.7 K using VTI cryostat unless mentioned.

The Landau fan diagram at zero D is shown in Fig. 1(b). The top and back gate voltages are converted to n and D using the estimated top and back gate capacitances (see Fig. 1(d)). We observe a number of LL crossings and asymmetry between the electron side and the hole side. These features have been reported in ABA-stacked TLG [24, 28], and are accounted for by the coexistence of MLG-like and BLG-like bands, which are gapped and energetically detuned from each other owing to the tight-binding parameters γ_2 , γ_5 , δ , and $\Delta_2 = \frac{1}{6}[(U_1 - U_2) - (U_2 - U_3)]$ defined by the onsite energy of each layer, U_1 , U_2 , and U_3 .

We numerically calculated the band structure around K point (left graph) and B dependence of the LLs energies at $D = 0$ V/nm (right graph) for comparison as shown in Fig. 1(c). The calculation was performed using the tight-binding model [22, 23] with the parameters, $\gamma_0 = 3.1$ eV, $\gamma_1 = 0.39$ eV, $\gamma_2 = -0.028$ eV, $\gamma_3 = 0.315$ eV, $\gamma_4 = 0.041$ eV, $\gamma_5 = 0.05$ eV and $\delta = 0.046$ eV adopted from Ref. [24]. Approximated wave functions of the LLs for the low energy at $D = 0$ V/nm are summarized in Table I. We define labeling of the LLs in the mirror-symmetric monolayer (m) and mirror-antisymmetric bilayer (b) by LL orbital basis. (Note $\frac{|A_1\rangle - |A_3\rangle}{\sqrt{2}}$ is mirror-symmetric owing to the antisymmetric property of π orbitals.) Their energies, derived analytically by neglecting the small correction by γ_3 , are shown in Table II for the small LL indices. The LL labels are shown visually in Fig. 1(c) (right).

By comparing Fig. 1(b) with Fig. 1(c), the LLs marked by the pentagon, hexagon, and stars in Fig. 1(b) are identified as $+LL3_{\pm}^b$, $-LL3_{\pm}^b$ and $-LL1_{\pm}^m$, respectively. The LLs marked by the triangle and circle in Fig. 1(b) are identified as either $LL0_{\pm}^m$ or $LL0_{\pm}^m$. The LL indicated by the square mark in Fig. 1(b) is identified as either $LL0_{\pm}^b$ or $LL1_{\pm}^b$. Depending on the tight-binding parameters such as γ_2 , γ_5 , δ , and Δ_2 , the order of the LL energies with small indices changes (see Table II)[31]. Therefore, we cannot uniquely identify the indices for the LLs indicated by the triangle, circle and square marks.

We then investigated the D dependence of the transport characteristics. D modifies the band structure and induces hybridization between the MLG-like and BLG-like bands. Fig. 1(d) shows the n and D dependences of the longitudinal resistance R_{xx} measured at 10K at $B = 0$ T. The n and D values were calculated using the

top and back gate capacitances, which were estimated from the slope of the R_{xx} peak position in the plane defined by the top and back gate voltages and the known parameters of the gate insulators: The thickness, and dielectric constant of SiO_2 is 285nm, and 3.9, respectively, and the thickness of the top, and bottom h-BN layers is 44.1nm, and 52.7nm, respectively. The R_{xx} peak at $n = 0$ increases as D increases. This is understood by the reduction of the density of states around the charge neutrality point (CNP) by the band hybridization due to the mirror symmetry breaking under D [22, 26]. Such band evolution with D , expected from the tight-binding model with full parameters [22, 31], is shown in Fig. 2(c).

Figs. 2(a), and 2(b) show the n and D dependences of R_{xx} for $B = 2.8$ T, and 1.4 T, respectively. The R_{xx} measured along $D = 0$ V/nm in Figs. 2(a), and 2(b) corresponds to the R_{xx} measured along the broken green lines (i), and (ii) in Fig. 1(c), respectively. In Figs. 2(a) and 2(b), we see a number of LLs parallel to the D axis. By comparing them with Fig. 1(c) at $D = 0$ V/nm, most of these LLs, such as the LL marked by the pentagon ($+LL3_{\pm}^b$), are found to originate from the BLG-like bands. In addition, several sequences of LL crossings identified by the kinks of the parallel LLs are observed in Figs. 2(a) and 2(b). We put eye guides over some of the LLs which show characteristic evolution with D leading to LL crossings. As shown in Fig. 2(c), with increasing D , the MLG-like bands move away from each other while the BLG-like bands gradually overlap due to the band hybridization. It is therefore expected that most of the LLs in the MLG-like bands move away from the charge neutrality point (CNP) while most of the LLs in the BLG-like bands gradually approach the CNP.

These trends are clearly observed in Fig. 2(b). By following the LLs indicated by the eye guides shown in red and comparing them with those in Fig. 1(b) at $D = 0$ V/nm, it is found that these LLs originate from the MLG-like bands. One of the LLs indicated by the eye guides shown in blue quickly moves away from the CNP with D . However, in comparison with Fig. 1(b), this LL is found to originate from the BLG-like band.

The detailed evolution of LLs under D is understood by considering the sublattice degree of freedom of LL wave functions summarized in Table I and their hybridization. Note that $|n\rangle$ is the LL orbital with index n . D affects Δ_1 , which is half of the energy detuning between the top and bottom layers. Under D , $\frac{|A_1\rangle - |A_3\rangle}{\sqrt{2}}$ (monolayer) and $\frac{|A_1\rangle + |A_3\rangle}{\sqrt{2}}$ (bilayer) of the same $|n\rangle$ and valley hybridize. From Table I, we can identify four kinds of such hybridization processes:

- (H1) $LL0_{\pm}^m$ and $LL0_{\pm}^b$
- (H2) $LL1_{\pm}^b$ and $\pm LL1_{\pm}^m$
- (H3) $\pm LLn_{\pm}^m$ and $\pm LLn_{\pm}^b$ ($n \geq 2$)
- (H4) $\pm LL(n-1)_{\pm}^m$ and $\pm LLn_{\pm}^b$ ($n \geq 2$).

We schematically show these processes in Fig. 2(d). In

TABLE I: The correspondence between LL names and their wave function at $D = 0$ V/nm. We adopted the notation of the LLs introduced by Ref. [31]. The numbering of the LLs is based on the dominant and larger LL orbital indices. The superscript “m” (“b”) indicates that the LL is the eigenstate of the monolayer (bilayer) block of the Hamiltonian. The subscript “+” (“-”) indicates valley K (K’). $+LLn_{\pm}^{m,b}$ has larger energy than $-LLn_{\pm}^{m,b}$. For the low energy, the wave function of bilayer block LLs are obtained from two band approximation of the original 4×4 Hamiltonian, owing to the high energy of $|A_2\rangle$ and $\frac{|B_1\rangle+|B_3\rangle}{\sqrt{2}}$. Here we neglected the trigonal warping term (γ_3). $a_n^{m,b\pm}, b_n^{m,b\pm}$ are non-zero coefficients.

LL	Wave function	LL	Wave function
$LL0_+^m$	$ 0\rangle \otimes \frac{ B_1\rangle- B_3\rangle}{\sqrt{2}} \otimes K\rangle$	$LL0_-^m$	$ 0\rangle \otimes \frac{ A_1\rangle- A_3\rangle}{\sqrt{2}} \otimes K'\rangle$
$LL0_+^b$	$ 0\rangle \otimes B_2\rangle \otimes K\rangle$	$LL0_-^b$	$ 0\rangle \otimes \frac{ A_1\rangle+ A_3\rangle}{\sqrt{2}} \otimes K'\rangle$
$LL1_+^b$	$ 1\rangle \otimes B_2\rangle \otimes K\rangle$	$LL1_-^b$	$ 1\rangle \otimes \frac{ A_1\rangle+ A_3\rangle}{\sqrt{2}} \otimes K'\rangle$
$+LLn_+^m$	$\left(a_n^{m+} n-1\rangle \otimes \frac{ A_1\rangle- A_3\rangle}{\sqrt{2}} + b_n^{m+} n\rangle \otimes \frac{ B_1\rangle- B_3\rangle}{\sqrt{2}}\right) \otimes K\rangle$	$+LLn_-^m$	$\left(a_n^{m-} n\rangle \otimes \frac{ A_1\rangle- A_3\rangle}{\sqrt{2}} + b_n^{m-} n-1\rangle \otimes \frac{ B_1\rangle- B_3\rangle}{\sqrt{2}}\right) \otimes K'\rangle$
$-LLn_+^m$	$\left((b_n^{m+})^* n-1\rangle \otimes \frac{ A_1\rangle- A_3\rangle}{\sqrt{2}} - (a_n^{m+})^* n\rangle \otimes \frac{ B_1\rangle- B_3\rangle}{\sqrt{2}}\right) \otimes K\rangle$	$-LLn_-^m$	$\left((b_n^{m-})^* n\rangle \otimes \frac{ A_1\rangle- A_3\rangle}{\sqrt{2}} - (a_n^{m-})^* n-1\rangle \otimes \frac{ B_1\rangle- B_3\rangle}{\sqrt{2}}\right) \otimes K'\rangle$
$+LLn_+^b$	$\left(a_n^{b+} n-2\rangle \otimes \frac{ A_1\rangle+ A_3\rangle}{\sqrt{2}} + b_n^{b+} n\rangle \otimes B_2\rangle\right) \otimes K\rangle$	$+LLn_-^b$	$\left(a_n^{b-} n\rangle \otimes \frac{ A_1\rangle+ A_3\rangle}{\sqrt{2}} + b_n^{b-} n-2\rangle \otimes B_2\rangle\right) \otimes K'\rangle$
$-LLn_+^b$	$\left((b_n^{b+})^* n-2\rangle \otimes \frac{ A_1\rangle+ A_3\rangle}{\sqrt{2}} - (a_n^{b+})^* n\rangle \otimes B_2\rangle\right) \otimes K\rangle$	$-LLn_-^b$	$\left((b_n^{b-})^* n\rangle \otimes \frac{ A_1\rangle+ A_3\rangle}{\sqrt{2}} - (a_n^{b-})^* n-2\rangle \otimes B_2\rangle\right) \otimes K'\rangle$

(H1), two LLs, $LL0_-^m$ and $LL0_-^b$, move away from each other. In the rest of (H2), (H3), and (H4), two LLs in the MLG-like bands and one or two LLs in the BLG-like bands hybridize. For not too small B , the LLs in the BLG-like bands usually stay between the two LLs in the MLG-like bands. Therefore, the two LLs in the MLG-like bands move away from each other for increasing D . On the other hand, the LLs in the BLG-like bands are repelled by them from both sides and slightly get closer to each other.

Note in the D range studied here, hybridization between $\frac{|B_1\rangle-|B_3\rangle}{\sqrt{2}}$ and $\frac{|B_1\rangle+|B_3\rangle}{\sqrt{2}}$ has a minor influence on the LLs owing to the large energy gap between them. Therefore the energy of $LL0_+^m$ is almost unchanged with D . $LL0_+^b$ and $LL1_+^b$, which are polarized to $|B_2\rangle$, are also expected to be almost independent of D .

In Fig. 2(c), we schematically show the expected evolution of the specific LLs with D . The top of the valence band of the BLG-like bands splits off into the top of the green-colored band and that of the magenta-colored

band owing to the hybridization with the valence band of the MLG-like bands. One of the hybridized bands then moves away towards the lower energy and forms a new valence band (green). This process leads to the observation that $LL0_-^b$ moves away with other LLs of the MLG-like bands with increasing D , since only the top of the green-colored band has the BLG-like $\frac{|A_1\rangle+|A_3\rangle}{\sqrt{2}}$ component and the rest originates from the MLG-like components. On the other hand, the top of the valence band of the MLG-like bands (top of the band colored with light blue) remains unchanged with D although there is hybridization with the BLG-like band. This leads to the observation in Fig. 2(a) that the energy of $LL0_+^m$ as well as those of LLs in the BLG-like bands remains almost constant.

From the above considerations, we identified that the LL marked by the triangle, circle, and square in Fig. 2(a) are $LL0_-^m$, $LL0_+^m$, and $LL0_-^b$, respectively. The hybridization process (H1) separates $LL0_-^m$ and $LL0_-^b$ with increasing D while $LL0_+^m$ stays at the constant energy owing to the weak influence of the $\frac{|B_1\rangle+|B_3\rangle}{\sqrt{2}}$ orbitals. Note that evolutions of these LLs are B independent (common in Figs. 2(a) and 2(b)) around $D = 0$. This further supports that these are the zeroth LLs, representing the edges of the bands as described in Fig. 2(c).

However, in Fig. 2(a) the eye guide (B) passes through $LL0_-^b$ at $D = 0$ V/nm, and becomes almost independent of D for $|D| > 0.2$ V/nm. In contrast, the eye guide (C), which passes through $-LL3_{\pm}^b$ at $D = 0$ V/nm, shifts away from $n \sim -0.7$ cm $^{-2}$ for $|D| > 0.2$ V/nm. The observed behavior around $|D| \sim 0.2$ V/nm is interpreted as the anti-crossing between $LL0_-^b$ and $-LL3_-^b$. It has been pointed out that the trigonal warping term γ_3 hybridizes every third LLs of the BLG-like bands [31]. This means that the LLs of the BLG-like bands which mainly consist of $|n\rangle$, are influenced by $|n \pm 3\rangle, |n \pm 6\rangle, \dots$. Therefore,

TABLE II: Analytically derived energies for each LLs at $D = 0$ V/nm by neglecting the trigonal warping term (γ_3). ζ is defined by $\zeta = 3eBa^2\gamma_0^2/(4\hbar\gamma_1^2)$, where a is lattice constant.

LL	Energy
$LL0_+^m$	$\delta - \frac{1}{2}\gamma_5 + \Delta_2$
$LL0_-^m$	$-\frac{1}{2}\gamma_2 + \Delta_2$
$LL0_+^b$	$-2\Delta_2$
$LL1_+^b$	$-2\Delta_2 + \zeta\left(\frac{\gamma_5}{2} + \delta + \Delta_2\right)$
$LL0_-^b$	$\frac{\gamma_2}{2} + \Delta_2$
$LL1_-^b$	$\frac{\gamma_2}{2} + \Delta_2 + \zeta(\delta - 2\Delta_2)$

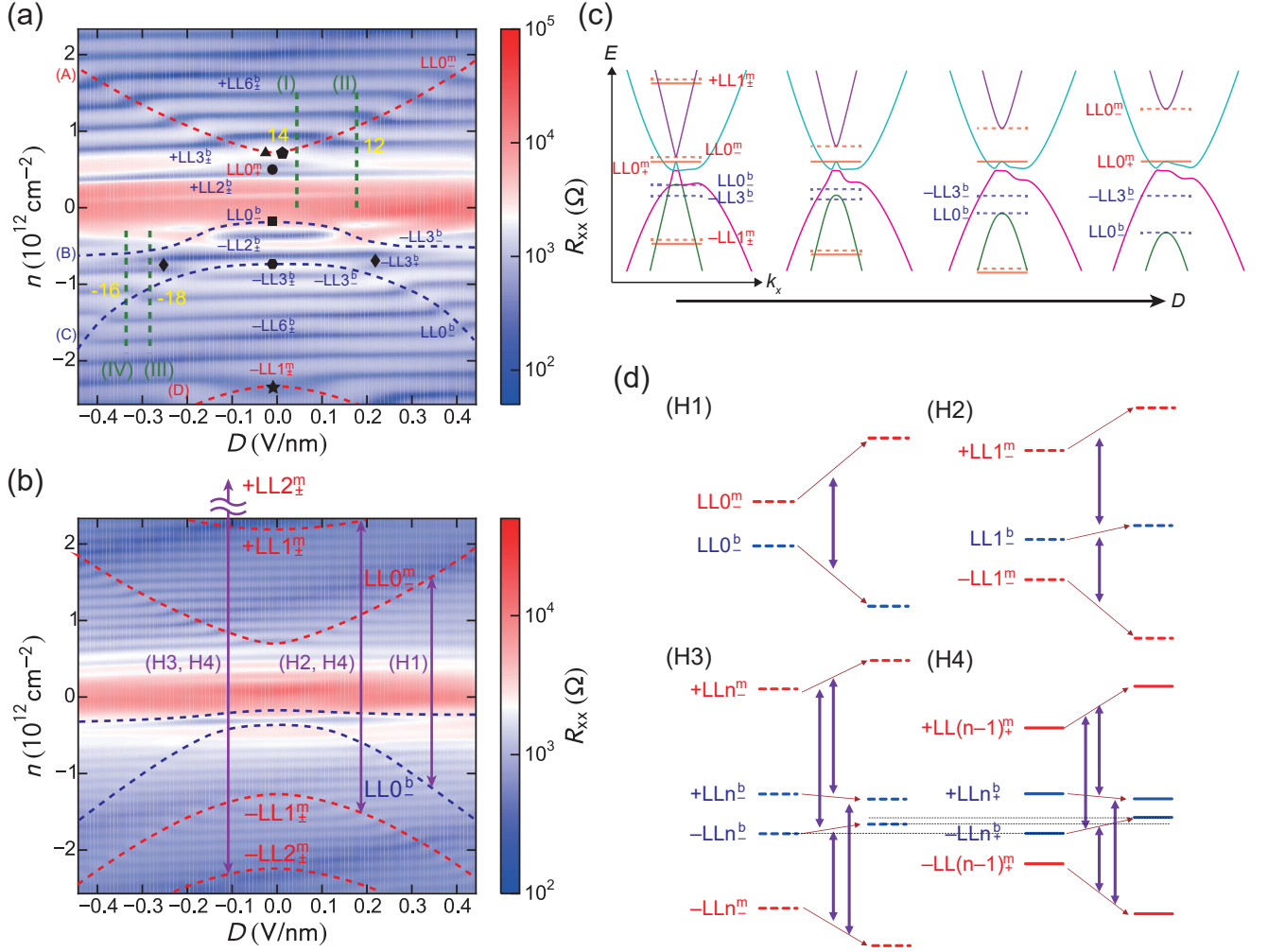


FIG. 2 (a) and (b) n and D the dependences of R_{xx} under $B = 2.8 \text{ T}$, and 1.4 T , respectively. The broken curves in red (blue) are eye guides, which follow the evolution of the specific LLs in the MLG-like (BLG-like) bands. The marks in black indicate the LLs, corresponding to the ones shown in Fig. 1(b) except for the LL marked by the diamonds. The numbers in yellow indicate the filling factors between the LLs. The purple double sided arrows indicate the LLs involved in each hybridization process. (c) Calculated band structure evolution with D . The LLs are schematically shown by the solid (broken) lines, which exist at valley K (K'). The following tight-binding parameters were used for the calculation of the band structure evolution :

$\gamma_0 = 3.1 \text{ eV}$, $\gamma_1 = 0.39 \text{ eV}$, $\gamma_2 = -0.028 \text{ eV}$, $\gamma_3 = 0.315 \text{ eV}$, $\gamma_4 = 0.041 \text{ eV}$, $\gamma_5 = 0.05 \text{ eV}$, $\delta = 0.034 \text{ eV}$, and $\Delta_2 = 0 \text{ eV}$. Δ_1 was modulated from 0 eV to 0.06 eV at regular intervals. (d) Schematic image of the hybridization processes with D . The purple double sided arrows indicate pairs of LLs which directly hybridize under D .

when we consider the evolution of $LL0_{\pm}^{\text{b}}$ and $LL0_{\pm}^{\text{m}}$ by the hybridization under D , $\pm LL3_{\pm}^{\text{m,b}}$, $\pm LL6_{\pm}^{\text{m,b}}$, \dots also have to be taken into account.

In contrast to $-LL3_{\pm}^{\text{b}}$, which shows anti-crossing with $LL0_{\pm}^{\text{b}}$, $-LL3_{\pm}^{\text{b}}$ remains unaffected. Therefore, $-LL3_{\pm}^{\text{b}}$, marked by the diamonds in Fig. 2(a), splits off from $-LL3_{\pm}^{\text{b}}$, crosses over the anti-crossing structure, and merges again with $-LL3_{\pm}^{\text{b}}$. Note this is the first observation of the trigonal warping mediated valley splitting.

To further confirm this scenario, we show numerical

calculations of LLs energies E_{LL} vs Δ_1 at $B = 2.8 \text{ T}$, for $\gamma_3 = 0.315 \text{ eV}$ (left graph) and $\gamma_3 = 0 \text{ eV}$ (right graph) in Fig. 3(a), where Δ_1 is half the energy detuning between the top and bottom layers, which is controlled by D . The calculation was based on Ref. [31]. Here, we used the same tight-binding parameters as used for the calculation of Fig. 1(c). The experimental result (Fig. 2(a)) including the anti-crossing described above is well reproduced in the left graph of Fig. 3(a). We clearly see the anti-crossing structure between $LL0_{\pm}^{\text{b}}$ and $-LL3_{\pm}^{\text{b}}$ for

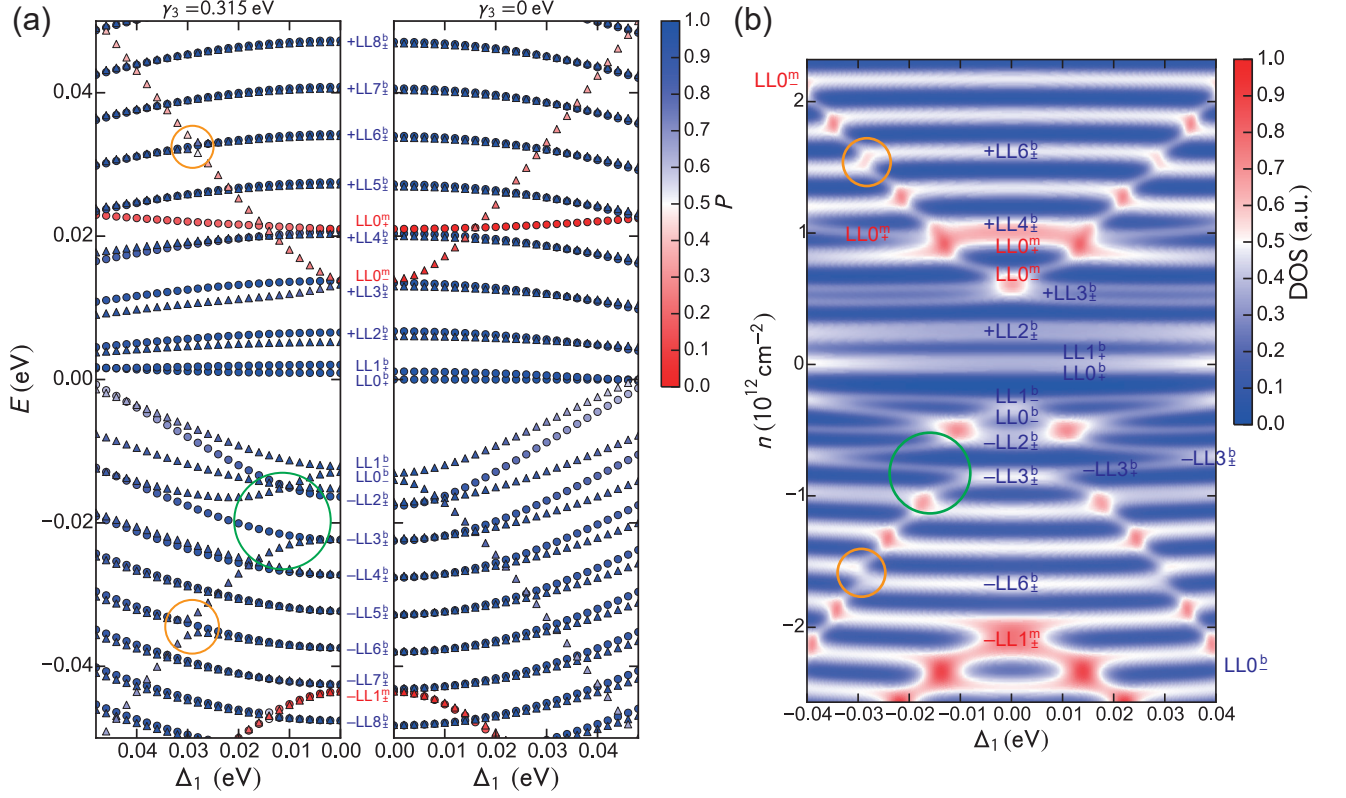


FIG. 3 (a) Numerically calculated Δ_1 dependence of the LLs energies at $B = 2.8$ T from the tight-binding model. The parameters assumed are the same as used in the calculation of Fig. 1(c) in the left graph, and the same except for $\gamma_3 = 0$ eV in the right graph. The colorscale indicates the portion of the wave function belonging: the wave function of the blue (red) points mainly belongs to the bilayer (monolayer) block. The circle (triangle) shaped points belong to K (K') valley. The green and orange circles indicate the anti-crossings between LLs due to the trigonal warping. (b) Numerically calculated n and Δ_1 dependences of the LLs DOS at $B = 2.8$ T from the tight-binding model. The parameters are the same as used in the calculation of Fig. 1(c). The green and orange circles indicate the anti-crossings between LLs due to the trigonal warping.

$\gamma_3 = 0.315$ eV (indicated by the green circle), but not for $\gamma_3 = 0$ eV. Though, we see small anti-crossing structures between $LL0^b_-$ and $-LL6^b_-$, $LL0^m$ and $+LL6^b_-$ for $\gamma_3 = 0.315$ eV in the calculation (indicated by the orange circles), they are not visible in Fig. 2(a). This is attributed to line broadening of the LLs. In Fig. 3(b), we show the Δ_1 and n dependences of the LLs density of states (DOS) calculated from the energy level E_{LL} in Fig. 3(a) (left graph). Here the DOS of each LL is assumed as: $DOS(E; E_{LL}) = \frac{2B}{h/e} \frac{1}{\pi} \frac{\Gamma/2}{(E - E_{LL})^2 + (\Gamma/2)^2}$ using the level broadening $\Gamma = 2$ meV, where h , and e are the Planck constant, and elementary charge, respectively [24]. We find good agreement of this calculation with the experimental result in Fig. 2(a). Note that the order of some LLs in Figs. 3(a) and 3(b) is different from that in Fig. 2(a). For example, the order of $LL0^m_+$ and $LL0^m_-$, which is determined by γ_2, γ_5 and δ (see Table II), is opposite. This is probably due to the environmental difference or the imperfection of the estimation of the tight binding pa-

rameters in previous work [24] in which Δ_2 is neglected.

The consistency of the LLs assignment between the experiment and calculation is further confirmed by investigating the degeneracy of the observed LLs. The LLs indicated by the eye guides (A) and (C) ($LL0^m_-$ and $-LL3^b_-$) in Fig. 2(a) are considered to be valley polarized to K'. Therefore, the filling factor should change by two across these lines. Fig. 4 shows the n dependence of the Hall conductivity σ_{xy} at different D simultaneously measured with R_{xx} in Fig. 2(a). (I) to (IV) indicate the correspondence between the green broken lines (I) to (IV) in Fig. 2(a). In Fig. 4(a), the filling factor around $n \sim 0.8 \times 10^{12} \text{ cm}^{-2}$ changes from 14 (left graph) to 12 (right graph). Also, in Fig. 4(b), the filling factor around $n \sim -1.2 \times 10^{12} \text{ cm}^{-2}$ changes from -18 (left graph) to -16 (right graph). These observations are consistent with the interpretation that the evolving LLs indicated by the eye guides (A) and (C) are valley polarized. In the left graph of Fig. 4(b), we see the change of the filling factor

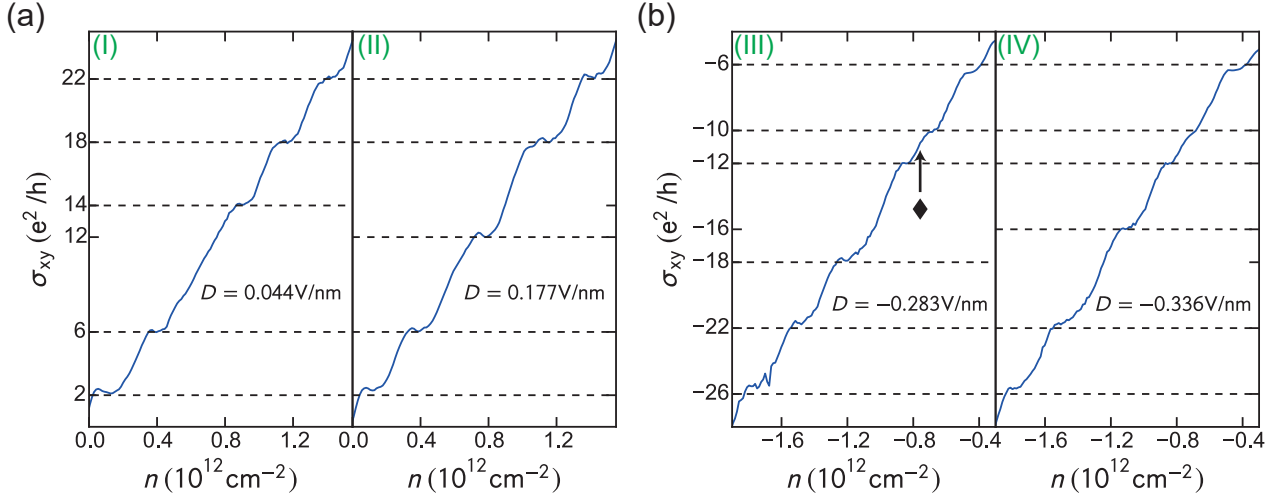


FIG. 4 (a) n dependence of σ_{xy} for each fixed D at $B = 2.8 \text{ T}$: $D = 0.044 \text{ V/nm}$ (I) and $D = 0.177 \text{ V/nm}$ (II). (b) n dependence of σ_{xy} for each fixed D at $B = 2.8 \text{ T}$: $D = -0.283 \text{ V/nm}$ (III) and $D = -0.336 \text{ V/nm}$ (IV). (I) to (IV) indicate σ_{xy} simultaneously measured with R_{xx} along the broken green lines of (I) to (IV) in Fig. 2(a).

from -12 to -10 around $n \sim -0.8 \times 10^{12} \text{ cm}^{-2}$ across the LL marked by the diamonds in Fig. 2(a). Therefore, this LL has two-fold degeneracy. This is also consistent with the interpretation that the anti-crossing between LL0^m and $-\text{LL3}^b$ results in the valley splitting of $-\text{LL3}^b$ and the LL marked by the diamonds is $-\text{LL3}^b$.

Finally, we show the LLs evolution with n and D at a high B of 5.6 T in Fig. 5(a). The R_{xx} measured along $D = 0 \text{ V/nm}$ in Fig. 5(a) corresponds to the R_{xx} measured along the broken green line (iii) in Fig. 1(c). At this B , many fine structures are visible, for instance spin splittings of LLs for LL0^m and LL0^b . Fig. 5(b) shows numerically calculated Δ_1 dependence of the LLs energies at $B = 5.6 \text{ T}$, using the same tight-binding parameters as in the calculation of Fig. 3(a). In Fig. 5(c), we show the Δ_1 and n dependences of the LLs DOS calculated in the same manner as used for the calculation of Fig. 3(b). The LLs evolution in Fig. 5(a) compares well to the calculation of Figs. 5(b) and 5(c). From this comparison, we identify the LLs evolution as indicated by the eye guides in Fig. 5(a).

Around $D \sim 0.1 \text{ V/nm}$, we see a kink structure in the evolution of LL0^b indicating the presence of an additional LL crossing. LL0^b and LL1^b are the possible crossing states, but γ_2 , γ_5 , δ , and Δ_2 (see Table II) should be carefully evaluated to judge between them.

In Fig. 5(a), in addition to the anti-crossing between LL0^b and $-\text{LL3}^b$, we also see anti-crossing between LL0^m and $+\text{LL3}^b$. This is in contrast with the case at $B = 2.8 \text{ T}$ where LL0^m and $+\text{LL3}^b$ degenerate but don't hybridize at $D = 0 \text{ V/nm}$ (see Fig. 2(a)). Under such a high B , $+\text{LL3}^b$ shifts to the high energy with B , leading to the these LLs crossings at non-zero D . This observa-

tion indicates that both the trigonal warping term (γ_3) that hybridizes LL0^b and $-\text{LL3}^b$ and the mirror symmetry breaking by D that hybridizes LL0^m and LL0^b (H1) are required to form the anti-crossing structure.

In Fig. 5(d), we magnified the region of the anti-crossing between LL0^m and $+\text{LL3}^b$ for $D < 0$. While the process of the anti-crossing, spin splitting of LL0^m disappears and recovers again across the anti-crossing. This observation is also consistent with anti-crossings, across which the LL index is preserved.

We also observe an effect of the hybridization (H3) and (H4) in Fig. 5(a). It is the valley splitting of $-\text{LL2}^b$ for $|D| > 0.3 \text{ V/nm}$. $-\text{LL2}^b$ mainly hybridizes with $-\text{LL1}^m$ through the process (H4) while $-\text{LL2}^b$ mainly hybridizes with $-\text{LL2}^m$ through (H3). Since the energy difference between $-\text{LL2}^b$ and $-\text{LL1}^m$ is smaller than that between $-\text{LL2}^b$ and $-\text{LL2}^m$, $-\text{LL2}^b - -\text{LL1}^m$ hybridization is stronger than $-\text{LL2}^b - -\text{LL2}^m$. This results in valley splitting.

Through out the paper we neglected intervalley hybridization and assumed that two valley states behave independently. The actual device has rough edges because it was shaped by dry etching and therefore may experience intervalley hybridization of the edge states. However, this will not affect the bulk LLs which we mainly discussed above. In the counter-propagating edge transport regime, electron and hole channels coexist and the channel number is influenced by hybridization of edge states. In the $\nu = 2$ regime at $B = 5.6 \text{ T}$ and $D = 0 \text{ V/nm}$ (indicated by a green arrow in Fig. 5(a)), usual two chiral edge transport ($R_{xx} \sim 0 \Omega$ and $R_{xy} \sim \frac{1}{2} \frac{h}{e^2}$) has been observed inspite of the expected counter-propagating edge transport which consists of four elec-

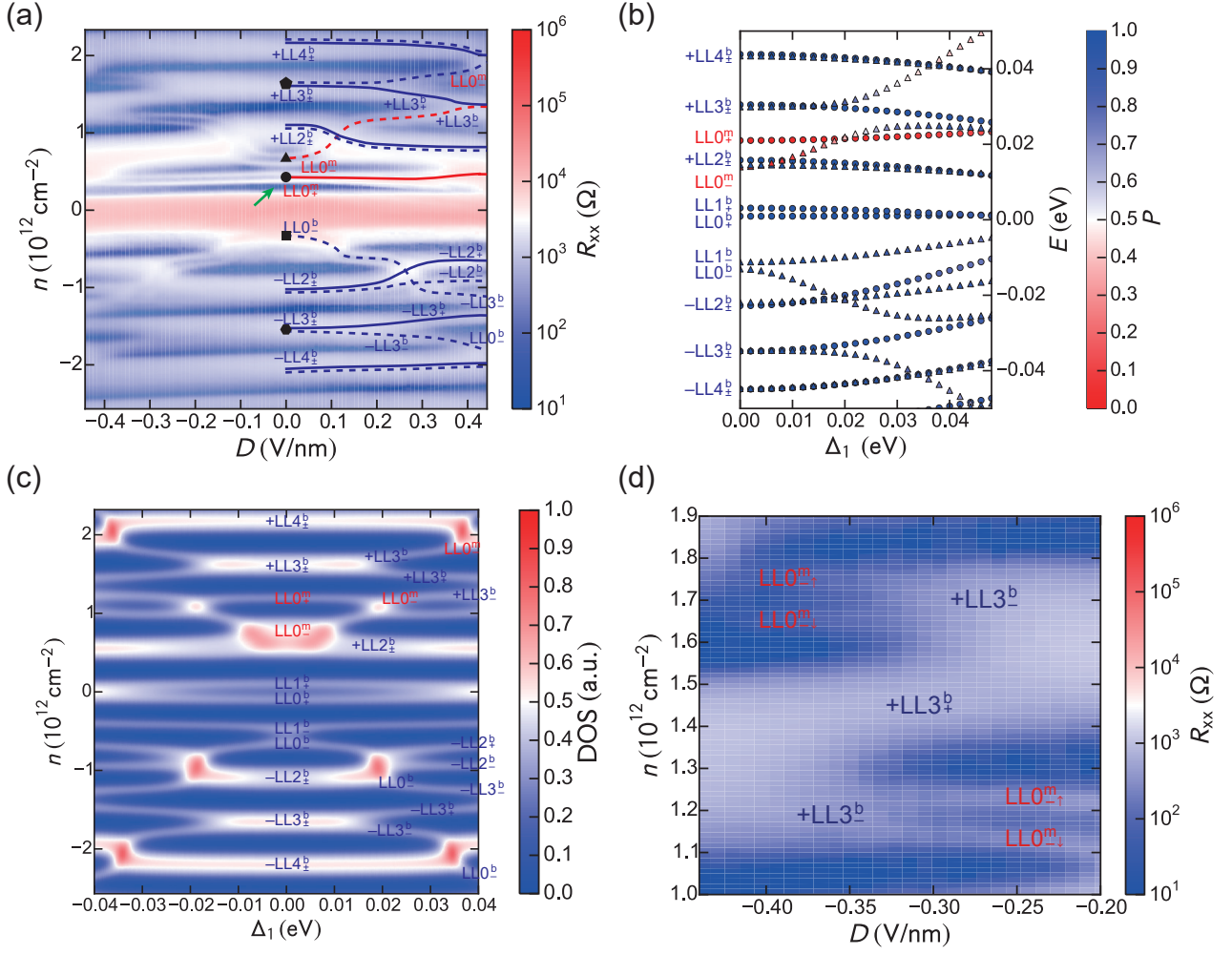


FIG. 5 (a) n and D dependences of R_{xx} measured at $B = 5.6$ T. The curves in red (blue) are eye guides, which follow the evolution of the specific LLs in the MLG-like (BLG-like) bands. The LLs indicated by the solid (broken) curves belong to K (K') valley. (b) Numerically calculated Δ_1 dependence of the LLs energies at $B = 5.6$ T. The parameters assumed are the same as used in the calculation of Fig. 1(c). The colorscale indicates the portion of the wave function belonging: the wave function of the blue (red) points mainly belongs to the bilayer (monolayer) block. The circle (triangle) shaped points belong to K (K') valley. (c) Numerically calculated n and Δ_1 dependences of the LLs DOS at $B = 5.6$ T. The parameters assumed are the same as used in the calculation of Fig. 1(c). (d) Extracted and magnified figure from the area around the anti-crossing between LL0_m^- and LL0_b^- .

tron channels and two hole channels. This can involve the intervalley hybridization of the edge states by rough edges where the total edge states are reduced to two chiral edge states.

In summary, we observed LL crossings under an electric field in ABA-stacked trilayer graphene, and find that the overview of the LL evolution is consistent with the theory of hybridized MLG-like and BLG-like bands. The observed LL evolution shows a variety of valley and orbital dependences. We find that it is explained by considering the hybridization between the multiple LLs sharing the same LL orbital and valley and the hybridization be-

tween every third LL orbitals due to the trigonal warping term γ_3 . We also find pronounced agreement between our experimental observations and numerical calculation based on the tight binding model [31]. We finally revealed the entire picture of the LL evolution with electric field in ABA-stacked trilayer graphene, and showed a variety of quantum Hall phase can be achieved via the mirror symmetry breaking using electric field. While preparing the manuscript, we found related works on the quantum Hall phases achieved in different regimes [35, 36].

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- [1] Y. Zhang, Y.-W. Tan, H. L. Stormer, and P. Kim, *Nature* **438**, 201 (2005).
- [2] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos, and A. A. Firsov, *Nature* **438**, 197 (2005).
- [3] A. F. Young, C. R. Dean, L. Wang, H. Ren, P. Cadden-Zimansky, K. Watanabe, T. Taniguchi, J. Hone, K. L. Shepard, and P. Kim, *Nat. Phys.* **8**, 550 (2012).
- [4] P. Maher, C. R. Dean, A. F. Young, T. Taniguchi, K. Watanabe, K. L. Shepard, J. Hone, and P. Kim, *Nat. Phys.* **9**, 154 (2013).
- [5] A. F. Young, J. D. Sanchez-Yamagishi, B. Hunt, S. H. Choi, K. Watanabe, T. Taniguchi, R. C. Ashoori, and P. Jarillo-Herrero, *Nature* **505**, 528 (2014).
- [6] B. Hunt, J. D. Sanchez-Yamagishi, A. F. Young, M. Yankowitz, B. J. LeRoy, K. Watanabe, T. Taniguchi, P. Moon, M. Koshino, P. Jarillo-Herrero, and R. C. Ashoori, *Science* **340**, 1427 (2013).
- [7] L. A. Ponomarenko, R. V. Gorbachev, G. L. Yu, D. C. Elias, R. Jalil, A. A. Patel, A. Mishchenko, A. S. Mayorov, C. R. Woods, J. R. Wallbank, M. Mucha-Kruczyński, B. A. Piot, M. Potemski, I. V. Grigorieva, K. S. Novoselov, F. Guinea, V. I. Fal’ko, and A. K. Geim, *Nature* **497**, 594 (2013).
- [8] C. R. Dean, L. Wang, P. Maher, C. Forsythe, F. Ghahari, Y. Gao, J. Katoch, M. Ishigami, P. Moon, M. Koshino, T. Taniguchi, K. Watanabe, K. L. Shepard, J. Hone, and P. Kim, *Nature* **497**, 598 (2013).
- [9] J. B. Oostinga, H. B. Heersche, X. Liu, A. F. Morpurgo, and L. M. K. Vandersypen, *Nat. Mater.* **7**, 151 (2007).
- [10] Y. Zhang, T.-T. Tang, C. Girit, Z. Hao, M. C. Martin, A. Zettl, M. F. Crommie, Y. R. Shen, and F. Wang, *Nature* **459**, 820 (2009).
- [11] H. Shioya, M. Yamamoto, S. Russo, M. F. Craciun, and S. Tarucha, *Appl. Phys. Lett.* **100**, 033113 (2012).
- [12] C. H. Lui, Z. Li, K. F. Mak, E. Cappelluti, and T. F. Heinz, *Nat. Phys.* **7**, 944 (2011).
- [13] T. Khodkov, I. Khrapach, M. F. Craciun, and S. Russo, *Nano Lett.* **15**, 4429 (2015).
- [14] R. T. Weitz, M. T. Allen, B. E. Feldman, J. Martin, and A. Yacoby, *Science* **330**, 812 (2010).
- [15] S. Kim, K. Lee, and E. Tutuc, *Phys. Rev. Lett.* **107**, 016803 (2011).
- [16] J. Velasco Jr., L. Jing, W. Bao, Y. Lee, P. Kratz, V. Aji, M. Bockrath, C. N. Lau, C. Varma, R. Stillwell, D. Smirnov, F. Zhang, J. Jung, and A. H. MacDonald, *Nat. Nanotechnol.* **7**, 156 (2012).
- [17] P. Maher, L. Wang, Y. Gao, C. Forsythe, T. Taniguchi, K. Watanabe, D. Abanin, Z. Papić, P. Cadden-Zimansky, J. Hone, P. Kim, and C. R. Dean, *Science* **345**, 61 (2014).
- [18] K. Lee, B. Fallahazad, J. Xue, D. C. Dillen, K. Kim, T. Taniguchi, K. Watanabe, and E. Tutuc, *Science* **345**, 58 (2014).
- [19] J. Velasco Jr., Y. Lee, F. Zhang, K. Myhro, D. Tran, M. Deo, D. Smirnov, A. H. MacDonald, and C. N. Lau, *Nat. Commun.* **5**, 4550 (2014).
- [20] Y. Lee, D. Tran, K. Myhro, N. Gillgren, J. M. Poumirol, D. Smirnov, Y. Barlas, and C. N. Lau, *Nano Lett.* **16**, 227 (2016).
- [21] Y. Shi, Y. Lee, S. Che, Z. Pi, T. Espiritu, P. Stepanov, D. Smirnov, C. N. Lau, and F. Zhang, *Phys. Rev. Lett.* **116**, 056601 (2016).
- [22] M. Koshino and E. McCann, *Phys. Rev. B* **79**, 125443 (2009).
- [23] M. Koshino and E. McCann, *Phys. Rev. B* **83**, 165443 (2011).
- [24] T. Taychatanapat, K. Watanabe, T. Taniguchi, and P. Jarillo-Herrero, *Nat. Phys.* **7**, 621 (2011).
- [25] M. F. Craciun, S. Russo, M. Yamamoto, J. B. Oostinga, A. F. Morpurgo, and S. Tarucha, *Nat. Nanotechnol.* **4**, 383 (2009).
- [26] E. A. Henriksen, D. Nandi, and J. P. Eisenstein, *Phys. Rev. X* **2**, 011004 (2012).
- [27] K. Zou, F. Zhang, C. Clapp, A. H. MacDonald, and J. Zhu, *Nano Lett.* **13**, 369 (2013).
- [28] Y. Lee, J. Velasco, D. Tran, F. Zhang, W. Bao, L. Jing, K. Myhro, D. Smirnov, and C. N. Lau, *Nano Lett.* **13**, 1627 (2013).
- [29] M. Koshino and E. McCann, *Phys. Rev. B* **81**, 115315 (2010).
- [30] T. Morimoto and M. Koshino, *Phys. Rev. B* **87**, 085424 (2013).
- [31] M. Serbyn and D. A. Abanin, *Phys. Rev. B* **87**, 115422 (2013).
- [32] A. Varlet, D. Bischoff, P. Simonet, K. Watanabe, T. Taniguchi, T. Ihn, K. Ensslin, M. Mucha-Kruczyński, and V. I. Fal’ko, *Phys. Rev. Lett.* **113**, 116602 (2014).
- [33] S. H. Jhang, M. F. Craciun, S. Schmidmeier, S. Tokumitsu, S. Russo, M. Yamamoto, Y. Skourski, J. Wosnitza, S. Tarucha, J. Eroms, and C. Strunk, *Phys. Rev. B* **84**, 161408 (2011).
- [34] L. Wang, I. Meric, P. Y. Huang, Q. Gao, Y. Gao, H. Tran, T. Taniguchi, K. Watanabe, L. M. Campos, D. A. Muller, J. Guo, P. Kim, J. Hone, K. L. Shepard, and C. R. Dean, *Science* **342**, 614 (2013).
- [35] L. C. Campos, T. Taychatanapat, M. Serbyn, K. Surakitbovorn, K. Watanabe, T. Taniguchi, D. A. Abanin, and P. Jarillo-Herrero, *Phys. Rev. Lett.* **117**, 066601 (2016).
- [36] P. Stepanov, Y. Barlas, T. Espiritu, S. Che, K. Watanabe, T. Taniguchi, D. Smirnov, and C. N. Lau, *Phys. Rev. Lett.* **117**, 076807 (2016).